Modelling Hysteresis and Irreversibility in Magnetic Materials:
A Two-Level Subsystem Approach

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“Hysteresis is at the heart of the behaviour of magnetic materials. All applications, from electric motors to transformers and permanent magnets, from various types of electronic devices to magnetic recording, rely heavily on particular aspects of hysteresis. It is a beautiful example of a physical and mathematical problem of intriguing elegance and challenging complexity that is at the same time the source of pervading technological progress.”

G. Bertotti

*Hysteresis in Magnetism, Academic Press*
The Magnetic Response of a Magnetically "Frozen" Material Has Two Principal Components:

**Critical Component:**
- dominates the response of the material in the vicinity of the critical temperature $T_c$, and is directly related to the spontaneous, cooperative magnetic “condensation” driven by the quantum mechanical exchange interaction between the atomic moments.

**Irreversible, History-Dependent Component:**
- dominates the magnetic response of the material at essentially all temperatures below $T_c$, and is related to the tendency for the material to form coarse-spatial-scale metastable magnetic structures in an effort to minimize the total free energy, which includes orientational, anisotropic, and surface contributions.
I have studied a wide variety of magnetic materials including ferromagnets and nano-particulate assemblies.

\[(x=0.5) \quad \text{La}_{x}\text{Sr}_{1-x}\text{CoO}_3 \quad \text{Polycrystalline ferromagnets, with perovskite structure} \]

\[(x=0.2,0.4,0.6) \quad \text{Ca}_{x}\text{Sr}_{1-x}\text{RuO}_3 \quad \text{Fe nanoparticles embedded in insulating matrix} \]

\[
\begin{align*}
\text{Fe/Al}_2\text{O}_3 \\
\text{Fe/SiO}_2 \\
\text{Magnetite (Fe}_3\text{O}_4) \\
\text{(x=0.1) Titanomagnetite (Fe}_{3-x}\text{Ti}_x\text{O}_4) \\
\end{align*}
\]

\[\text{Nanodimensional grains} \quad \text{Nanodimensional grains embedded in volcanic glass} \]

These materials are characterized by considerable natural structural disorder.
Experimentally, the irreversible response of a magnetic material is characterized by measuring the following standard macroscopic functions as a function of applied field $H_a$, temperature $T$, and observation time $t$:

- Field Cooled (FC) Moment
- Zero Field Cooled (ZFC) Moment
- ThermoRemanent Moment (TRM)
- Isothermal Remanent Moment (IRM)
- Hysteresis Loops
- Relaxation Isotherms
FC/ZFC Moment

FC moment - measured upon warming after cooling in a field from a temperature in the reversible regime

ZFC moment - measured upon warming after cooling in a field and applying a field

Nano-magnetite

Polycrystalline Ca$_{0.4}$Sr$_{0.6}$RuO$_3$
Hysteresis Loops

Remanent Hysteresis Loops

Temperature Dependence of Coercive Field

NanoFe in alumina

Polycrystalline $Ca_x Sr_{1-x} RuO_3$
Relaxation Isotherms

NanoMagnetite

Moment as a function of time at constant temperature after recoiling from positive saturation to a negative holding field $-H_a$ in the vicinity of the coercive field $H_c$

NanoFe embedded in alumina

Nano-magnetite
Preisach Collections of Bistable Elements

- Multi-valley free energy landscape is characterized by a large number of local minima, maxima, and saddle points

- Preisach postulate: the free energy landscape can be decomposed into an ensemble of elementary two-level subsystems

\[ W_- = W_d + W_s \]
\[ W_+ = W_d - W_s \]

or

\[ W_d = \frac{W_- + W_+}{2} \]
\[ W_s = \frac{W_- - W_+}{2} \]

\[ W_s = +\mu H_s \]
\[ W_d = +\mu H_d \]
Thermal activation is described by applying the Arrhenius law for the time required to jump over a barrier of height $W$.

This defines the maximum barrier which can be activated thermally:

$$\dot{\omega} = \dot{\omega}_0 \exp \left( \frac{W}{k_B T} \right)$$

$$W_T = k_B T \ln(t_{\text{exp}}/\dot{\omega}_0)$$

The free energy profile of a two-level Preisach subsystem in a positive applied field with thermal activation.
Current Issues

a) Determination of the Thermal Fluctuation Field

- Among the most challenging problems facing any Preisach reconstruction of the metastable state excitation spectrum is the determination of the thermal fluctuation field \( H_f = kT/\mu \).
- The Preisach formalism suggests a method for extracting the thermal fluctuation field based on an analysis of viscosity isotherms.

Thermal fluctuation field as a function of temperature for nanoFe embedded in alumina.

**Magnon Condensation?**

**Or**

**Interaction Effects?**
Aging and Memory Effects

- Experimental signatures of collective spin glass freezing include i) slow relaxation dynamics, extending from microscopic to geological time scales, and ii) aging, which refers to the dependence of the relaxation response on the time for which the system is held at constant temperature after cooling from the paramagnetic state.

However, Preisach collections of two-level subsystems also exhibit aging and memory effects, and consequently it is essential to establish which of the anomalous relaxation effects observed experimentally are genuinely collective in origin and which are a simple product of thermal activation over individual, single-particle energy barriers.
Aging and Memory Effect
Experiments on Fe/Al$_2$O$_3$

i) Memory Steps

ii) Relaxation of ZFC moment interrupted by negative temperature cycling

Preisach Simulations

i) Memory Steps

ii) Relaxation of ZFC moment interrupted by negative temperature cycling
Further Issues to Contemplate

1) Explain the rich diversity observed in the temperature dependence of the measured coercive field.

2) How are interaction effects manifested in the macroscopic response functions?

3) How do we distinguish local interactions from collective interactions which result in cooperative freezing of the nanoparticle moments? (aging and memory effects?)

[Graphs: nanoFe in glass and Preisach]